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TITLE: Pb MIGRATION IN THE OKLO URANIUM DEPOSIT

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Pb MIGRATION IN THE OKLO URANIUM DEPOSIT. A. J. Gancarz and . **B. Curtis**, University of California, Los Alamos Scientific Laboratory, Los Alamos, New Mexico 87545

Walton and Cowan (1) and Bryant *et al.* (2) suggested that the natural fission reactors discovered at the Oklo uranium deposit in Gabon, Africa and the geologic environment around the reactors offered an opportunity to study the redistribution of elements on a geologic time scale. The setting is unique because the reactors produced a suite of elements with distinct isotopic abundances and rocks external to the reactor which contain these isotopically anomalous elements, represent zones into which reactor products migrated. In addition, reactor nucleonics calculations quantitatively predict the amounts of the reactor products; thus, losses of these elements from reactor zones are easily detected. Finally, many of the reactor products and the U itself are radioactive and as clocks can be used to determine *rates* of element migration.

Uranium-lead studies of Oklo U-ore samples, both from within and external to the reactor zone, show that samples are depleted in Pb relative to the amounts calculated from U decay (3,4). The average loss is ~50%. Gancarz (5) interpreted the data to indicate a primary age of 2.05 $\times$ 10^9 years with Pb loss from uraninite by continuous volume diffusion. We report U-Pb and Pb isotopic data for additional ore and non-ore samples selected in an attempt to delineate the paths along which the lost Pb migrated. These latter samples were chosen from a large number of core samples, both above and below the uranium rich zones. They are all from a conglomeratic layer 3 to 5 meters underneath the U-ore and reactor zones. Rocks from this layer were the only ones to contain either micro or macroscopic crystals of pyrite (FeS_2) and/or galena (PbS_2).

Shown on Figure 1 are the new data and those previously reported by Gancarz (5). The non-ore samples are those with $^{238}U/^{206}Pb^* < 0.5$. The heavy curve is the concordia curve, i.e., the locus of points for closed system Pb evolution from pure U between the time indicated on the curve and the present. Data to the right of the curve indicate Pb loss relative to U and data to the left reflect relative Pb gain. The curve through the data is a model curve. It is the locus of points assuming a starting time of 2.05 AE and continuous loss of Pb by

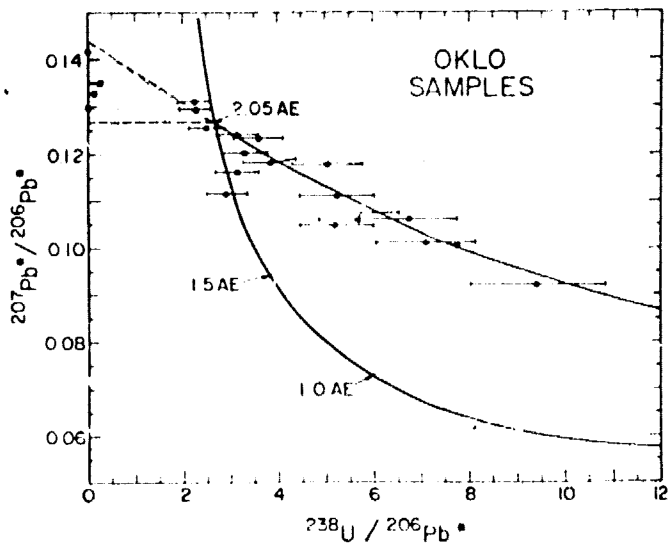

FIGURE 1

volume diffusion for various values of the effective diffusion coefficient, D/a^2 (6). The hatched area is where data plot for samples which have lost no Pb and gained diffused Pb. We conclude from these data that Pb is lost from host uraninite by a diffusion mechanism. The conglomerate underlying the U deposit contains this diffused Pb and is, therefore, both a transport path and repository for Pb. The significance of the variation of $^{207}Pb^*/^{206}Pb^*$ in the conglomerate remains unclear as does to why data plot below the diffusion trajectory.

The Pb incorporated into rocks at the time of their formation has been subtracted from the data before they are plotted on Figure 1. For data plotted on Figure 2 this initial component is not subtracted. The circles represent rocks depleted in Pb as deduced from Figure 1 and the squares are for the Pb enriched samples. The curve labeled $\hat{\mu}$ is the Pb diffusion curve for $^{238}U(today)/^{204}Pb(initial)$ = 10 000. The close agreement of the data to this model curve suggests that not only *in situ* uranogenic Pb, but also the *initial* Pb is lost by diffusion. The two parallel lines, one labeled "isochron," define the region where samples which have retained Pb and have gained diffused Pb plot. As with the U-Pb systematics, these Pb isotopic data alone show that the conglomerate contains

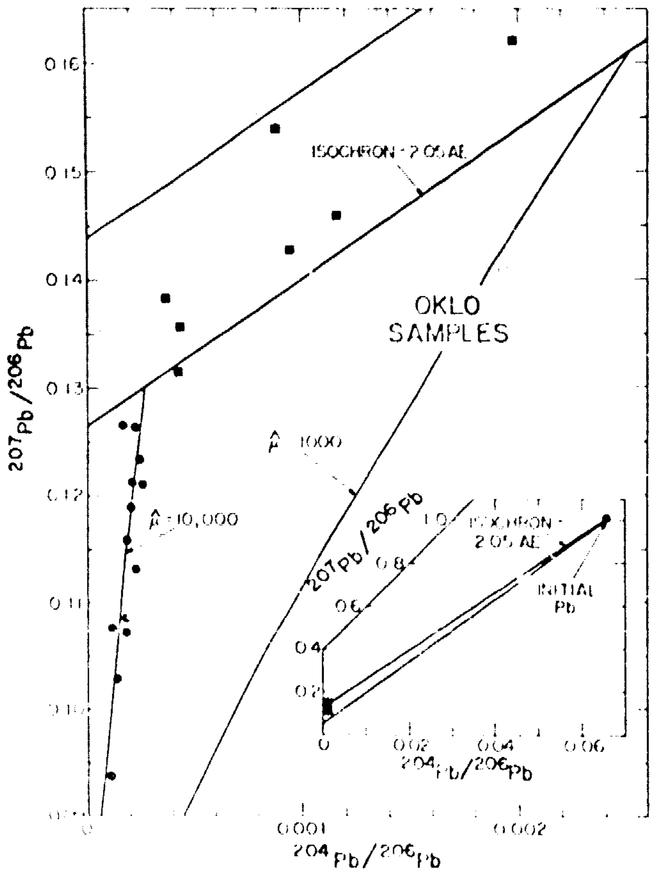

FIGURE 2

exce.ss Pb, which is the component diffused from the uraninite, and is a transport zone and repository for the Pb.

These U-Pb data demonstrate the potential of the Oklo uranium ore deposit and natural fission reactors as a long time scale analogue for man-made radioactive waste repositories. The extent to which other reactor products migrated through specific zones and/or were retained in these zones needs yet to be determined.

REFERENCES

1. Walton, R. D. and Cowan, G. A. (1975) in: *The Oklo Phenomenon* I.A.E.A., Vienna, p. 499.

2. Bryant, E. A., Cowan, G. A., Daniels, W. R., and Maeck, W. J. (1976) in: *Actinides In the Environment* A.C.S. Symp. Series No. 35, p. 89.

3. Lancelot, J. R., Vitrac, A., and Allegre, C. J. (1975) Earth Planet. Sci. Lett. 25, p. 189.

4. Gauthier-Lafaye, F., Besnus, Y., and Weber, F. (1978) in: *Natural Fission Reactors*, I.A.E.A., Vienna, p. 35.

5. Gancarz, A. J. *ibid.*, p. 513.

6. Tilton, G. R. (1960) J. Geophys. Res. 65, p. 2933.